

FORM PTO-1390 (REV 5-93)		U.S. DEPARTMENT OF COMMERCE PATENT AND TRADEMARK OFFICE		ATTORNEY'S DOCKET NUMBER	
TRANSMITTAL LETTER TO THE UNITED STATES DESIGNATED/ELECTED OFFICE (DO/EO/US) CONCERNING A FILING UNDER 35 U.S.C. 371				1748X/49135	
				U.S. APPLICATION NO. (if known, see 37 CFR 1.5) 09/623011	
INTERNATIONAL APPLICATION NO. PCT/EP99/01144		INTERNATIONAL FILING DATE February 23, 1999		PRIORITY DATE CLAIMED February 25, 1998	
TITLE OF INVENTION FUEL CELL SYSTEM					
APPLICANT(S) FOR DO/EO/US Arnold LAMM, Jens MÜLLER, Norbert WIESHEU					
Applicant herewith submits to the United States Designated/Elected Office (DO/EO/US) the following items and other information:					
<p>1. <input checked="" type="checkbox"/> This is a FIRST submission of items concerning a filing under 35 U.S.C. 371.</p> <p>2. <input type="checkbox"/> This is a SECOND or SUBSEQUENT submission of items concerning a filing under 35 U.S.C. 371</p> <p>3. <input checked="" type="checkbox"/> This express request to begin national examination procedures (35 U.S.C. 371(f) at any time rather than delay examination until the expiration of the applicable time limit set in 35 U.S.C. 371(b) and PCT Articles 22 and 39(1).</p> <p>4. <input checked="" type="checkbox"/> A proper Demand for International Preliminary Examination was made by the 19th month from the earliest claimed priority date.</p> <p>5. <input checked="" type="checkbox"/> A copy of the International Application as filed (35 U.S.C. 371(c)(2)).</p> <p style="margin-left: 40px;">a. <input type="checkbox"/> is transmitted herewith (required only if not transmitted by the International Bureau).</p> <p style="margin-left: 40px;">b. <input checked="" type="checkbox"/> has been transmitted by the International Bureau</p> <p style="margin-left: 40px;">c. <input type="checkbox"/> is not required, as the application was filed in the United States Receiving Office (RO/US)</p> <p>6. <input checked="" type="checkbox"/> A translation of the International Application into English (35 U.S.C. 371(c)(2)).</p> <p>7. <input type="checkbox"/> Amendments to the claims of the International Application under PCT Article 19 (35 U.S.C. 371(c)(3)).</p> <p style="margin-left: 40px;">a. <input type="checkbox"/> are transmitted herewith (required only if not transmitted by the International Bureau).</p> <p style="margin-left: 40px;">b. <input type="checkbox"/> have been transmitted by the International Bureau.</p> <p style="margin-left: 40px;">c. <input type="checkbox"/> have not been made; however, the time limit for making such amendments has NOT expired.</p> <p style="margin-left: 40px;">d. <input type="checkbox"/> have not been made and will not be made.</p> <p>8. <input type="checkbox"/> A translation of the amendments to the claims under PCT Article 19 (35 U.S.C. 371(c)(3)).</p> <p>9. <input checked="" type="checkbox"/> An oath or declaration of the inventor(s) (35 U.S.C. 371(c)(4)). UNEXECUTED</p> <p>10. <input checked="" type="checkbox"/> A translation of the annexes to the International Preliminary Examination Report under PCT Article 36 (35 U.S.C. 371(c)(5)).</p> <p>Item 11. to 16. below concern other document(s) or information included:</p> <p>11. <input checked="" type="checkbox"/> An Information Disclosure Statement under 37 CFR 1.97 and 1.98.</p> <p>12. <input type="checkbox"/> An assignment document for recording. A separate cover sheet in compliance with 37 CFR 3.28 and 3.31 is included.</p> <p>13. <input checked="" type="checkbox"/> A FIRST preliminary amendment.</p> <p style="margin-left: 40px;"><input type="checkbox"/> A SECOND or SUBSEQUENT preliminary amendment.</p> <p>14. <input type="checkbox"/> A substitute specification.</p> <p>15. <input type="checkbox"/> A change of power of attorney and/or address letter.</p> <p>16. <input checked="" type="checkbox"/> Other items or information:</p> <p style="margin-left: 40px;">PCT/IPEA/416</p> <p style="margin-left: 40px;">PCT/IB/308</p> <p style="margin-left: 40px;">cover page of international application</p> <p style="margin-left: 40px;">copy of International Search Report - PCT/ISA/210</p>					

U.S. APPLICATION NO (if known, see 37 CFR 1.55) 09/623011		INTERNATIONAL APPLICATION NO PCT/EP99/01144		ATTORNEY'S DOCKET NUMBER 1748X/49135	
17. <input checked="" type="checkbox"/> The following fees are submitted: Basic National Fee (37 CFR 1.492(a)(1)-(5)):				CALCULATIONS	
Search Report has been prepared by the EPO or JPO \$840.00 International preliminary examination fee paid to USPTO (37 CFR 1.482) ... \$670.00 No international preliminary examination fee paid to USPTO (37 CFR 1.482) but international search fee paid to USPTO (37 CFR 1.445(a)(2)) \$760.00 Neither international preliminary examination fee (37 CFR 1.482) nor international search fee (37 CFR 1.445(a)(2)) paid to USPTO \$970.00 International preliminary examination fee paid to USPTO (37 CFR 1.482) and all claims satisfied provisions of PCT Article 33(2)-(4) \$96.00					
ENTER APPROPRIATE BASIC FEE AMOUNT =				\$ 840.00	
Surcharge of \$130.00 for furnishing the oath or declaration later than <input type="checkbox"/> 20 <input checked="" type="checkbox"/> 30 months from the earliest claimed priority date (37 CFR 1.492(e)).				\$ 130.00	
Claims	Number Filed	Number Extra	Rate		
Total Claims	14-20=	00	X \$18.00	\$	
Independent Claims	5-3=	2	X \$78.00	\$ 156.00	
Multiple dependent claims(s) (if applicable)			+ \$260.00	\$	
TOTAL OF ABOVE CALCULATIONS =				\$ 1,126.00	
Reduction by 1/2 for filing by small entity, if applicable. Verified Small Entity statement must also be filed. (Note 37 CFR 1.9, 1.27, 1.28).				\$	
SUBTOTAL =				\$ 1,126.00	
Processing fee of \$130.00 for furnishing the English translation later than <input type="checkbox"/> 20 <input type="checkbox"/> 30 months from the earliest claimed priority date (37 CFR 1.492(f)).				\$	
TOTAL NATIONAL FEE =				\$ 1,126.00	
Fee for recording the enclosed assignment (37 CFR 1.21(h)). The assignment must be accompanied by an appropriate cover sheet (37 CFR 3.28,3.31). \$40.00 per property +				\$	
TOTAL FEE ENCLOSED =				\$ 1,126.00	
				Amount to be:	\$
				refunded	
				charged	\$
a. <input checked="" type="checkbox"/> A check in the amount of <u>\$1,126.00</u> for the filing is enclosed b. <input type="checkbox"/> Please charge my Deposit Account No. _____ in the amount of \$ _____ to cover the above fees. A duplicate copy of this sheet is enclosed. c. <input checked="" type="checkbox"/> The Commissioner is hereby authorized to charge any additional fees, which may be required, or credit any overpayment to Deposit Account No. <u>05-1323</u> . A duplicate copy of this sheet is enclosed.					
NOTE: Where an appropriate time limit under 37 CFR 1.494 or 1.495 has not been met, a petition to revive (37 CFR 1.137(a) or (b)) must be filed and granted to restore the application to pending status.					
SEND ALL CORRESPONDENCE TO: Evenson, McKeown, Edwards & Lenahan, P.L.L.C. 1200 G Street, N.W., Suite 700 Washington, D.C. 20005 Tel. No. (202) 628-8800 Fax No. (202) 628-8844					
				SIGNATURE DONALD D. EVENSON NAME 26,160 REGISTRATION NUMBER August 25, 2000 DATE	

09/623011
533 Rec'd PCT/PTO 25 AUG 2000

Attorney Docket: 1748X/49135
PATENT

IN THE UNITED STATES PATENT AND TRADEMARK OFFICE

Applicants: ARNOLD LAMM et al.

PCT NO.: PCT/EP99/01104

Serial No.: Not Assigned Yet Group Art Unit: TBA

Filed: August 25, 1999 Examiner: TBA

Title: LIQUID FUEL CELL SYSTEM

PRELIMINARY AMENDMENT

Box PCT
Assistant Commissioner for Patents
Washington, D.C. 20231

Sir:

Please enter the following amendments to the specification and claims prior to the examination of the application.

IN THE SPECIFICATION:

Page 1, before first line of text, insert --BACKGROUND AND SUMMARY OF THE INVENTION--; and line 29, change "budget" to --needed--.

Page 2, lines 22-26, change "To achieve this object, according to the invention a fuel cell system having the features of Claim 1 is proposed. As a result of inventive operation of the fuel cell involving water break-through from the anode compartment into the cathode compartment," to --In a preferred embodiment, the fuel cell system involves passing water through the anode compartment into the cathode compartment,--;

lines 33-34, delete "Further advantageous refinements of the invention are described in the dependent claims"; and

line 35, change "Advantageously" to --In a preferred method--.

Page 3, line 22, change "Advantageously" to --In a preferred embodiment--;

before line 30, insert --BRIEF DESCRIPTION OF THE DRAWINGS--; and.

before line 33, insert --DETAILED DESCRIPTION OF THE INVENTION--.

Page 4, line 31, change "so-called stack it", to --so-called stack, it--.

Page 7, line 18, change "breaking" to --passing--, and after "through", insert --the membrane 16--;

line 29, change "breakthrough" to --passthroungh--; and

line 30, change "through" to --of--.

Page 8, line 5, change "there and therefore be" to --there, and therefore, be--;

lines 9-10, change "formed there by the water-producing reaction proper, the oxidation of hydrogen." to --formed there by the oxidation of hydrogen--; and

line 35, change "the expander 32," to --the expander 32.--.

Page 9, line 1, change "advantageously, the" to --Thus, it is preferable that the--;

lines 8-16, "In the fuel cell 10, owing to operation in water-breakthrough mode and to the cooler normally

provided in the anode circuit being dispensed with, steady-state operation will therefore result at a temperature which, in addition to the positive pressure in the cathode compartment 14, on the one hand depends on the properties of the proton-conducting membrane 16 and, on the other hand, can also be set via the speed of the pump 34 which provides the volume flow on the anode side." to --In the fuel cell 10, owing to the operation in water-passthrough mode, a steady-state operating temperature can be set without the need of the cooler normally provided in the anode circuit. The steady-state operating temperature can be set by controlling the positive pressure in the cathode compartment 14 and/or the speed of the pump 34 which provides the volume flow on the anode side.--;

line 29, after "condensing out" insert --water--;

line 30, delete "the water which is lacking"; and

line 31, delete "described".

Page 10, line 1, change "Claims" to --WHAT IS CLAIMED IS:--.

IN THE CLAIMS:

Kindly cancel claims 1-9 on the Amended Sheets, and substitute therefor, new claims 10-23 as follows:

- 10. A fuel cell system, comprising:
 - 1) at least one fuel cell which has
 - a) an anode compartment,
 - b) a cathode compartment, and
 - c) a proton-conducting membrane which separates said anode compartment from said

cathode compartment and is capable of allowing water to pass;

2) a cathode circuit in which said cathode compartment is disposed and said cathode circuit further includes a cathode feeder for delivering oxygen-containing gas to said cathode compartment; and

3) an anode circuit in which said anode compartment is disposed and further includes a gas separator, an anode feeder for delivering a liquid coolant/fuel mixture to said anode compartment, and a pump for pumping the liquid coolant/fuel mixture to said anode compartment,

wherein cooling the coolant/fuel mixture circulating in the anode circuit is effected by the fuel cell which is designed for an operation involving water passing through said membrane from the anode compartment into the cathode compartment, and in that an operating temperature of the fuel cell is set by controlling pressure of said cathode compartment or the delivery of the liquid coolant/fuel mixture from said pump.

11. The fuel cell system of Claim 10, further comprising a expander unit disposed in said cathode circuit, wherein water vapour generated in the cathode compartment is delivered to said expander unit.

12. The fuel cell system of Claim 10, further comprising a compressor unit disposed in said cathode feeder.

13. The fuel cell system of Claim 11, further comprising a compressor unit disposed in said cathode feeder.

14. The fuel cell system of Claim 13, further comprising a supercharger intercooler, a cooler, and at least one water separator for water recovery, wherein said supercharger intercooler is disposed downstream of the compressor unit, and said cooler and at least one water separator are disposed downstream of the expander unit.

15. The fuel cell system of Claim 10, further comprising a holding and purification tank disposed in said anode circuit.

16. The fuel cell system of Claim 15, further comprising an anode offtake and a subsidiary branch of the anode offtake, wherein said holding and purification tank is disposed in said subsidiary branch upstream of said gas separator.

17. The fuel cell system of Claim 14, further comprising a feedback line, wherein recycling of recovered water from the at least one water separator into the anode circuit is provided via said feedback line.

18. A fuel cell system, comprising:

- 1) at least one fuel cell which has
 - a) an anode compartment,
 - b) a cathode compartment, and
 - c) a proton-conducting membrane which separates said anode compartment from said cathode compartment and is capable of allowing water to pass;

2) a cathode circuit in which said cathode compartment is disposed and said cathode circuit further includes a cathode feeder for delivering oxygen-containing gas to said cathode compartment; and

3) an anode circuit in which said anode compartment is disposed and further includes a gas separator, an anode feeder for delivering a liquid coolant/fuel mixture to said anode compartment, and a pump for pumping the liquid coolant/fuel mixture to said anode compartment,

wherein cooling the coolant/fuel mixture circulating in the anode circuit is effected by the fuel cell which is designed for an operation involving water passing through said membrane from the anode compartment into the cathode compartment, and in that an operating temperature of the fuel cell is set by controlling pressure of said cathode compartment and the delivery of the liquid coolant/fuel mixture from said pump.

19. A method of operating a fuel cell system having at least one fuel cell which includes an anode compartment and a cathode compartment which are separated from one another by a proton-conducting membrane, and an anode feeder for delivering a liquid coolant/fuel mixture to the anode compartment, comprising:

setting the operating temperature of the fuel cell by controlling pressure of the cathode compartment or a volume flow of the coolant/fuel mixture into the anode compartment;

passing water through the proton-conducting membrane from the anode compartment into the cathode compartment; and

cooling the coolant/fuel mixture in the anode compartment.

20. The method of claim 19, wherein the operating temperature is between 90 and 110°C.

21. A method of operating a fuel cell system having at least one fuel cell which includes an anode compartment and a cathode compartment which are separated from one another by a proton-conducting membrane, and an anode feeder for delivering a liquid coolant/fuel mixture to the anode compartment, comprising:

setting the operating temperature of the fuel cell by controlling pressure of the cathode compartment and a volume flow of the coolant/fuel mixture into the anode compartment;

passing water through the proton-conducting membrane from the anode compartment into the cathode compartment; and

cooling the coolant/fuel mixture in the anode compartment.

22. The method of claim 21, wherein the operating temperature is between 90 and 110°C.

23. A method of cooling a coolant/fuel mixture provided to a fuel cell system having at least one fuel cell that includes an anode compartment and a cathode compartment which are

separated from one another by a proton-conducting membrane, and an anode feeder for delivering a liquid coolant/fuel mixture to the anode compartment, comprising:

passing water through the proton-conducting membrane from the anode compartment into the cathode compartment; and

evaporating the water passing into the cathode compartment, whereby the evaporation of the water cools the coolant/fuel mixture in the anode compartment.--

REMARKS

Entry of the amendments to the specification and claims before examination of the application is respectfully requested. These claims have been amended to remove multiple dependencies and to put the claims in better form for prosecution. These claims patentably define over the art of record.

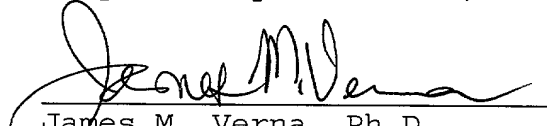
If there are any questions regarding this Preliminary Amendment or this application in general, a telephone call to the undersigned would be appreciated since this should expedite the prosecution of the application for all concerned.

It is respectfully requested that, if necessary to effect a timely response, this paper be considered as a Petition for an Extension of Time sufficient to effect a timely response and shortages in other fees, be charged, or any overpayment in fees

be credited, to the Account of Evenson, McKeown, Edwards & Lenahan, P.L.L.C., Deposit Account No. 05-1323 (Docket #1748X/49135).

August 25, 2000

Respectfully submitted,


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Original version

DBB Full Cell Engines GmbH

and

Ballard Power Systems Inc.

5

Fuel cell system

The invention relates to a fuel cell system comprising a fuel cell which includes an anode compartment and a cathode compartment which are separated from one another by a proton-conducting membrane.

At present, the method most widely envisaged for converting liquid energy sources into electrical energy in a fuel cell system comprising a proton exchange membrane (PEM fuel cell) all over the world is that of reforming methanol in a gas generation system. This involves a water/methanol mixture being evaporated and being converted, in a reformer, into hydrogen, carbon dioxide and carbon monoxide. Evaporation and reforming are very expensive in terms of the energy balance. This entails reduced efficiencies for the system as a whole. Moreover, gas beneficiation steps are required to clean the reforming gas. The cleaned gas is delivered to the PEM fuel cell system. Additionally, a cooler must be provided to cool the coolant/fuel mixture circulating in the anode circuit.

A further problem is that of the water used in the reforming process. The product water produced on the cathode side does not suffice to cover the water budget. Consequently, a separate water tank is required.

A so-called direct-methanol fuel cell system, as disclosed by US Patent 5 599 638, makes use of an aqueous methanol solution which reacts on the anode side to form carbon dioxide. The fuel cell system described there includes a so-called stack consisting of a plurality of interconnected fuel cells. The anode compartment of the stack forms part of an anode circuit, comprising a heat

exchanger to cool the coolant/fuel mixture which is ducted off from the anode outlet and contains carbon dioxide, a circulation tank in which the cooled mixture is added to a freshly supplied coolant/fuel mixture, a
5 gas separator which is integrated within the circulation tank and has the purpose of separating carbon dioxide, and a pump to feed the coolant/fuel mixture from the circulation tank into the anode compartment via a corresponding feeder. The oxygen- and water vapour-
10 comprising cathode off-gas of the known fuel cell system is passed through a water separator, the separated water being fed to the coolant/fuel mixture which is to be delivered to the anode circuit, and part of the remaining oxygen being passed to the oxidant supply for the cathode
15 compartment.

Based on this, it is an object of the invention to provide a simpler-design, compact fuel cell system comprising a proton-conducting membrane and having an improved overall efficiency.

20 To achieve this object, according to the invention a fuel cell system having the features of Claim 1 is proposed. As a result of inventive operation of the fuel cell involving water break-through from the anode compartment into the cathode compartment,
25 evaporation cooling is effected in the fuel cell as the water is absorbed by the hot air of the cathode compartment, said evaporation cooling being utilized according to the invention to cool the anode circuit. Owing to this measure, the cooler which otherwise has to
30 be provided in the anode circuit can be dispensed with.

Further advantageous refinements of the invention are described in the dependent claims.

Advantageously, the fuel cell is operated in heat balance equilibrium, i.e. the fuel cell is operated in a
35 steady state at a temperature which, on the one hand, depends on the properties of the proton-conducting membrane and, on the other hand, can be adjusted via the

speed of the liquid pump. Depending on the duty point, the temperature of the steady state operation is between 90 and 110°C. Setting a steady-state operating temperature is of crucial importance in increasing the efficiency of the fuel cell or of the stack formed from a plurality of fuel cells, since this will enable isothermal operation of the stack, i.e. temperature differences over the length of the stack of an order of magnitude of about 10°C, which are standard in known systems, will no longer occur, or only to an insignificant extent.

The inventive evaporation cooling in the fuel cell has the additional advantage that the mass flow of the dry air is increased by a factor of 1.5 to 2, entailing an increase in expander capacity by the same factor. This also entails energy savings for air supply in full-load operation.

Advantageously, an air cooler downstream of the expander is provided which is thermally coupled to the vehicle radiator and which serves for condensing out water to achieve a positive water balance in the system.

The invention is depicted schematically in the drawing with reference to a specific embodiment and is explained below in more detail with reference to the drawing.

The only figure shows a schematic depiction of the basic configuration of a fuel cell system according to the invention.

The fuel cell system depicted in the figure comprises a fuel cell 10 which consists of an anode compartment 12 and a cathode compartment 14, which are separated from one another by a proton-conducting membrane 16. Via an anode feeder 18, the anode compartment 12 is supplied with a liquid coolant/fuel mixture. The fuel used in this context can be any electrochemically oxidizable substance having the general structural formula $H-[-CH_2O-]_n-Y$, where $1 \leq n \leq 5$ and $Y=H$ or

Y=CH₃. The fuel cell system of the specific example shown is operated with liquid methanol as a fuel and water as a coolant. Even though the following is restricted to a description of the use of a water/methanol mixture, the scope of the present application is not meant to be limited to this specific example. Potentially suitable coolants include, in particular, liquids or ionic or nonionic additives to water which have good antifreeze properties. Possible fuels include, for example, branched variations on the abovementioned general formula, for example di- or trimethoxymethane.

An oxygen-containing gas is passed into the cathode compartment 14 via a cathode feeder 20. According to the specific example shown, ambient air is used for this purpose. In the fuel cell 10, the fuel is oxidized at the anode and the oxygen from the air is reduced at the cathode. For this purpose, the proton-conducting membrane 16 is coated with suitable catalysts on the appropriate surfaces. Protons are now able to migrate from the anode side through the proton-conducting membrane 16 and combine, at the cathode side, with the oxygen ions to form water. This electrochemical reaction gives rise to a voltage between the two electrodes. By connecting many such cells in parallel or in series to form a so-called stack it is possible to achieve voltages and current intensities which are sufficiently high to drive a vehicle.

Formed as a product at the anode outlet is a carbon dioxide gas enriched with water and methanol. This liquid/gas mixture is discharged from the anode compartment 12 by an anode offtake 22. The cathode exhaust air containing residual oxygen and water vapour is ducted off via a cathode off-gas line 24. To achieve good efficiency, the ambient air is provided at positive pressure in the cathode compartment 14. For this purpose, there is disposed in the cathode feeder 20 a compressor 28 driven by an electric motor 26 and with a supercharger

intercooler 29 downstream thereof, which compressor draws in the desired air mass flow and compresses it to the required pressure level. In the case of operation based on ambient air, an air filter 30 is preferably additionally provided in the inlet area of the cathode feeder 20 upstream of the compressor 28. Part of the energy required to compress the ambient air can be recovered with the aid of an expander 32 disposed in the cathode off-gas line 24. Preferably, the compressor 28, the expander 32 and the electric motor 26 are disposed on a common shaft. Control of the fuel cell output is achieved by open- or closed-loop control of the compressor speed and consequently of the available air mass flow.

On the anode side, the water/methanol mixture is circulated at a predefined pressure with the aid of a pump 34, so that an excess supply of fuel will be ensured at the anode at all times. The ratio of water to methanol in the anode feeder 18 is set with the aid of a sensor 36 which measures the methanol concentration in the anode feeder 18. Depending on this sensor signal, the concentration of the water/methanol mixture is then controlled, the liquid methanol being delivered from a methanol tank 38 via a methanol delivery line 40 and being injected into the anode feeder 18 with the aid of an injection nozzle 44 not shown in any detail. The injection pressure is generated by an injection pump 42 disposed in the methanol delivery line 40. The anode compartment 12 is therefore supplied at all times with a water/methanol mixture having a constant methanol concentration.

Then the carbon dioxide enriched with methanol vapour and water vapour must be separated from the liquid/gas mixture ducted off via the anode offtake 22. To this end, the liquid/gas mixture is delivered, via the anode offtake 22, to a gas separator 52 in which the carbon dioxide is separated off. The water/methanol

mixture remaining in the gas separator 52 is recycled into the anode feeder 18 via a line 54.

The humid carbon dioxide gas separated off in the gas separator 52 is cooled to as low a temperature as possible in a cooler 56, further methanol and water being condensed out in a downstream water separator 58. The remaining dry carbon dioxide with a small residual level of methanol is passed, via a line 60, to the cathode gas offtake 24, where it is mixed with the oxygen-rich cathode exhaust air.

To separate as much liquid water as possible from the cathode exhaust air, a first water separator 59 is provided downstream of the outlet of the cathode compartment 14, and a further water separator 61 is provided downstream of the expander 32, as much as possible of the water vapour formed on the cathode side being delivered to the expander 32. In this arrangement, the expander 32 serves as a compact condensing turbine at whose outlet part of the water vapour condenses out. The water collected in the water separators 59, 61 is then recycled, via a feedback line 64 with an integrated feedback pump 62, into a holding and purification tank 50 of a subsidiary branch 48, 66 of the anode circuit. In particular, the holding and purification tank 50 is an ion exchanger.

Provided in the anode circuit, downstream of the anode outlet in the anode offtake 22, is a branch line 48 which runs to the holding and purification tank 50. The outlet of the holding and purification tank 50 is again connected to the anode offtake 22, via a line 66 with an integrated valve 68, upstream of the gas separator 52. The holding and purification tank 50 serves to hold and to purify the water/methanol mixture from the anode compartment 12, the water separated in the water separator 58, and the product water produced on the cathode side and recycled into the anode circuit via the feedback line 64. The valve 68 firstly serves to prevent

reverse flow from the anode offtake 22 into the line 66, and secondly to establish that fraction of the mixture from the anode offtake 22 which is to be passed through the holding and purification tank.

5 According to the invention, the fuel cell 10 is operated with water breaking through from the anode compartment 12 into the cathode compartment 14. The liquid water thus reaching the cathode compartment 14 is partially absorbed as vapour, up to saturation limit, by
10 the dry, hot air entering the cathode compartment 14 via the cathode feeder 20. This results in evaporation cooling in the fuel cell 10, said evaporation cooling being utilized according to the invention to cool the coolant/fuel mixture circulating in the anode circuit.
15 Thus the cooler which is otherwise normally provided in the anode offtake 22 can be dispensed with.

 The water breakthrough is due to an electro-osmotic transport phenomenon through the membrane 16. On the anode side, water molecules cluster around each
20 proton. Electro-osmotic pressure causes the latter to migrate through the ion channels of the membrane 16, e.g. Nafion®, to the cathode side. The number of the bound water molecules in this situation is slightly temperature-dependent and also depends on the ion channel
25 diameter of the membrane 16. The higher the electro-osmotic transport coefficient of the membrane 16, the more water will reach the cathode side, be able to evaporate there and therefore be able to be utilized for evaporation cooling of the fuel cell 10.

30 The transport via the membrane 16 causes about ten times more water to pass into the cathode compartment 14 than is formed there by the water-producing reaction proper, the oxidation of hydrogen. In the case of e.g. a Nafion membrane, about 5 water molecules are bound to a
35 proton which migrates through the membrane 16, whereas only one water molecule per two protons is formed in the oxidation. At 80°C, on average slightly fewer than 5,

and at 120°C slightly more than 5 water molecules are bound to a proton. In the case of a membrane material having larger ion channels, more water molecules can be bound to a proton, fewer in the case of a membrane material having smaller ion channels.

The water passing through the membrane evaporates on the cathode side and cools the fuel cell by evaporation cooling.

Preferably, the temperature of the cathode is close to the boiling point of water, to evaporate as much of the permeating water as possible, the positive pressure prevailing at the cathode 14 being capable of being set in a simple manner to control the boiling point of water. At a positive pressure of 1 bar, the boiling point is about 120°C instead of 100°C at atmospheric pressure. The temperature of the fuel cell is established in accordance with the positive pressure applied at the cathode side.

The water vapour is delivered to the expander 32. It is particularly advantageous to prevent water vapour from condensing out en route to the expander 32; advantageously, the lines are thermally insulated in a suitable manner, to prevent the water vapour from condensing out. Equally, it is expedient to make allowances, regarding the connection lines between cathode 16 and expander 32, for the larger volume required for the water vapour by making the line diameters sufficiently large.

In the fuel cell 10, owing to operation in water-breakthrough mode and to the cooler normally provided in the anode circuit being dispensed with, steady-state operation will therefore result at a temperature which, in addition to the positive pressure in the cathode compartment 14, on the one hand depends on the properties of the proton-conducting membrane 16 and, on the other hand, can also be set via the speed of the pump 34 which provides the volume flow on the anode side.

Advantageously, the steady-state operating temperature is between 90 and 110°C, particularly 105°C. This allows the fuel cell or a stack formed of a plurality of fuel cells to be operated virtually isothermally.

- 5 Evaporation cooling additionally, as already mentioned above, has the advantage of increasing the mass flow of the dry air by a factor of from 1.5 to 2. Thus the capacity of the expander 32 is increased by the same factor, entailing energy savings for the air supply.
- 10 These savings are about 8 kW in full-load operation. An air cooler 46 disposed downstream of the expander 32 is thermally coupled to the vehicle radiator (not shown in any detail) and has the purpose of condensing out from the exhaust air stream the water which is lacking to
- 15 achieve a positive water balance in the system described.

DBB Full Cell Engines GmbH
and
Ballard Power Systems Inc.

FTP/S - MH
10.01.2000

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Claims

1. Fuel cell system comprising at least one fuel cell (10) which includes an anode compartment (12) and a cathode compartment (14) which are separated from one another by a proton-conducting membrane (16), further comprising a cathode feeder (20) to delivering oxygen-containing gas to the cathode compartment (14), an anode feeder (18) for delivering a liquid coolant/fuel mixture to the anode compartment (12), the anode compartment (12) being disposed in an anode circuit which comprises a gas separator and a pump (34), characterized in that cooling of the coolant/fuel mixture circulating in the anode circuit is effected by the fuel cell (10) which is designed for operation involving water break-through from the anode compartment (12) into the cathode compartment (14), and in that the operating temperature of the fuel cell (10) is set by controlling the cathode compartment (14) pressure and/or the delivery of the pump (34) in the anode circuit.

2. Fuel cell system according to Claim 1, characterized in that the water vapour generated in the cathode compartment (14) is essentially delivered to an expander unit (32).

3. Fuel cell system according to Claim 1, characterized in that the anode circuit comprises a holding and purification tank (50).

4. Fuel cell system according to Claim 3, characterized in that the holding and purification tank (50) is disposed in a subsidiary branch (48, 66) of the anode offtake upstream of the gas separator (52).

5. Fuel cell system according to any one of Claims 1 to 4, characterized in that the cathode compartment (14) is disposed in a cathode circuit comprising a compressor/expander unit (28, 32).

5 6. Fuel cell system according to Claim 5, characterized in that in the cathode circuit downstream of the compressor (28) a supercharger intercooler (29) and downstream of the expander (32) a cooler (46) and at least one water separator (61) for water recovery are
10 provided.

7. Fuel cell system according to Claim 6, characterized in that recycling of recovered water into the anode circuit is provided via a feedback line (64).

8. Fuel cell system according to Claim 7,
15 characterized in that recycling of recovered water into the holding and purification tank (50) is effected.

9. Method of operating a fuel cell system comprising at least one fuel cell (10) which includes an anode compartment (12) and a cathode compartment (14) which are
20 separated from one another by a proton-conducting membrane (16), further comprising an anode feeder (18) for delivering a liquid coolant/fuel mixture to the anode compartment (12),
characterized in that

25 operation of the proton-conducting membrane (16) involves water break-through from the anode compartment (12) into the cathode compartment (14), and cooling of the coolant/fuel mixture circulating in the anode circuit is effected by the fuel cell (10), the operating temperature
30 of the fuel cell (10) being set by controlling the cathode compartment (14) pressure and/or the volume flow of the coolant/fuel mixture into the anode compartment.

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DBB Full Cell Engines GmbH

and

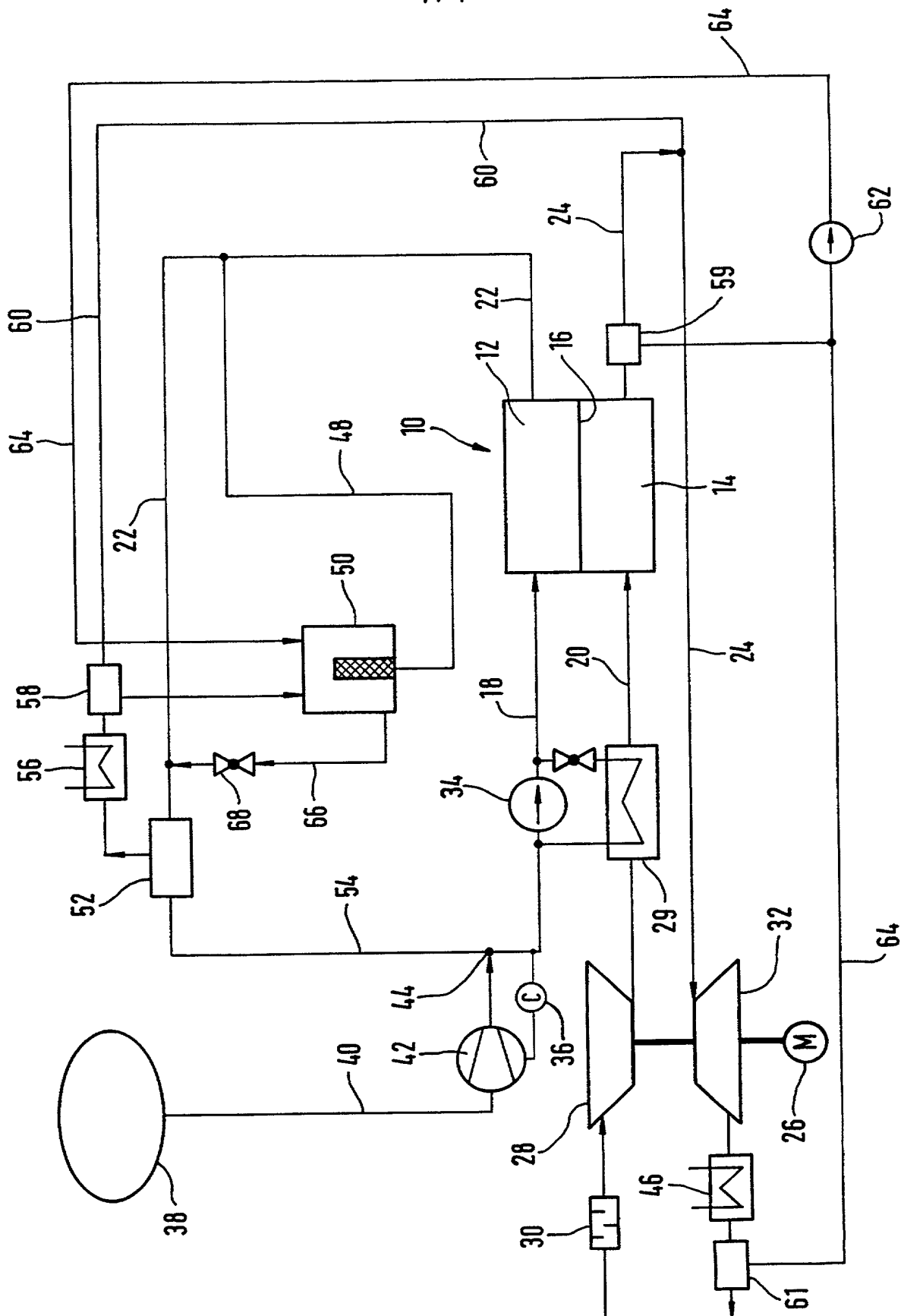
Ballard Power Systems Inc.

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Abstract

Fuel cell system comprising at least one fuel cell which includes an anode compartment and a cathode compartment which are separated from one another by a proton-conducting membrane, further comprising a cathode feeder for delivering oxygen-containing gas to the cathode compartment, an anode feeder for delivering a liquid coolant/fuel mixture to the anode compartment, the anode compartment being disposed in an anode circuit which comprises a gas separator and a pump, and cooling of the coolant/fuel mixture circulating in the anode circuit is effected by the fuel cell which is designed for operation involving water break-through from the anode compartment into the cathode compartment. The evaporation cooling thus achieved in the fuel cell results in cooling of the coolant/fuel mixture at a steady-state operating temperature which is established in the fuel cell as a function of the membrane properties and the speed of the pump, thus obviating the need for any additional cooler in the anode circuit itself.

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COMBINED DECLARATION FOR PATENT APPLICATION AND POWER OF ATTORNEY
(includes Reference to PCT International Applications)

ATTORNEY'S DOCKET NUMBER

1748X/49135

As a below named inventor, I hereby declare that:

My residence, post office address and citizenship are as stated below next to my name.

I believe I am the original, first and sole inventor (if only one name is listed below) or an original, first and joint inventor (if plural names are listed below) of the subject matter which is claimed and for which a patent is sought on the invention entitled:

FUEL CELL SYSTEM

the specification of which (check only one item below):

☐ is attached hereto.

☐ was filed as United States application

Serial No. _____
on _____
and was amended
on _____ (if applicable).

☒ was filed as PCT international application

Number PCT/EP99/01144
on February 23, 1999
and was amended under PCT Article 19
on _____ (if applicable).

I hereby state that I have reviewed and understand the contents of the above-identified specification, including the claims, as amended by any amendment referred to above.

I acknowledge the duty to disclose information which is material to the examination of this application in accordance with Title 37, Code of Federal Regulations. §1.56(a).

I hereby claim foreign priority benefits under Title 35, United State Code, §119 of any foreign application(s) for patent or inventor's certificate or of any PCT international application(s) designating at least one country other than the United States of America listed below and have also identified below any foreign application(s) for patent or inventor's certificate or any PCT international application(s) designating at least one country other than the United States of America filed by me on the same subject matter having a filing date before that of the application(s) of which priority is claimed:

PRIOR FOREIGN/PCT APPLICATION(S) AND ANY PRIORITY CLAIMS UNDER 35 U.S.C. 119:

COUNTRY (if PCT indicate PCT)	APPLICATION NUMBER	DATE OF FILING (day, month, year)	PRIORITY CLAIMED UNDER 35 USC 119
Germany	198 07 876.5	February 25, 1998	<input checked="" type="checkbox"/> Yes <input type="checkbox"/> No
			<input type="checkbox"/> Yes <input type="checkbox"/> No
			<input type="checkbox"/> Yes <input type="checkbox"/> No
			<input type="checkbox"/> Yes <input type="checkbox"/> No
			<input type="checkbox"/> Yes <input type="checkbox"/> No

Combined Declaration For Patent Application and Power of Attorney (Continued) (includes Reference to PCT international Applications)				ATTORNEY'S DOCKET NUMBER 1748X/49135	
I hereby claim the benefit under Title 35, United States Code, §120 of any United States application(s) or PCT international application(s) designating the United States of America that is/are listed below and, insofar as the subject matter of each of the claims of this application is not disclosed in that/those prior application(s) in the manner provided by the first paragraph of Title 35, United States Code, §112, I acknowledge the duty to disclose material information as defined in Title 37, Code of Federal Regulations, §1.56(a) which occurred between the filing date of the prior application(s) and the national of PCT international filing date of this application:					
PRIOR U.S. APPLICATIONS OR PCT INTERNATIONAL APPLICATIONS DESIGNATING THE U.S. FOR BENEFIT UNDER 35 U.S.C. 120					
U.S. APPLICATIONS				STATUS (Check one)	
U.S. APPLICATION NUMBER	U.S. FILING DATE	PATENTED	PENDING	ABANDONED	
PCT APPLICATIONS DESIGNATING THE U.S.					
PCT APPLICATION NO	PCT FILING DATE	U.S. SERIAL NUMBERS ASSIGNED (IF ANY)			
<p>POWER OF ATTORNEY: As a named inventor, I hereby appoint the following attorney(s) and/or agent(s) to prosecute this application and transact all business in the Patent and Trademark Office connected therewith. (List name and registration number)</p> <div style="display: flex; align-items: flex-start;"> <div style="font-size: 3em; margin-right: 10px; line-height: 1;">6</div> <div> <p>Herbert I. Cantor, Reg. No. 24,392; James F. McKeown, Reg. No. 25,406; Donald D. Evenson, Reg. No. 26,160; Joseph D. Evans, Reg. No. 26,269; Gary R. Edwards, Reg. No. 31,824; and Jeffrey D. Sanok, Reg. No. 32,169</p> </div> </div>					
Send Correspondence to:				Direct Telephone Calls to: (name and telephone number)	
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203	FULL NAME OF INVENTOR	FAMILY NAME <u>WIESHEU</u>	FIRST GIVEN NAME <u>NORBERT</u>	SECOND GIVEN NAME	
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	POST OFFICE ADDRESS	POST OFFICE ADDRESS <u>Georg-Lacher-Strasse 16</u>	CITY <u>D-89312 Günzburg</u>	STATE & ZIP CODE/COUNTRY Germany	
<p>I hereby declare that all statements made herein of my own knowledge are true and that all statements made on information and belief are believed to be true: and further that these statements were made with the knowledge that willful false statements and the like so made are punishable by fine or imprisonment, or both, under section 1001 of Title 18 of the United States Code, and that such willful false statements may jeopardize the validity of the application or any patent issuing thereon.</p>					
SIGNATURE OF INVENTOR 201 <i>[Signature]</i> <u>Lamm</u>		SIGNATURE OF INVENTOR 202 <i>[Signature]</i> <u>Müller</u>		SIGNATURE OF INVENTOR 203 <i>[Signature]</i> <u>Wiesher</u>	
DATE <u>28.8.2000</u>		DATE <u>28.8.2000</u>		DATE <u>28.8.2000</u>	